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Prediction of the amount of PCA for mechanical milling

Y.F. Zhang*, L. Lu, S.M. Yap

Department of Mechanical & Production Engineering, National University of Singapore, 10 Kent Ridge Crescent, Singapore 119260, Singapore

Abstract

Process control agent (PCA) can strongly influence the size of ball milled powder particles. Experimental results show that the mean particle size is affected by: (1) the types of the PCA, (2) the amount of PCA, and (3) the milling duration. Two kinds of materials, namely Al and Mg, were used in the experiment and analysis of the influence of process control agent. It was found that there is a critical amount of process control agent below which the size of the powder particles tends to increase and above which it tends to decrease. In order to predict the amount of PCA required for a particular mean particle size under a particular milling duration resulting from a particular mechanical alloying process, a back-propagation neural network is employed. For each combination of base material and PCA, a neural network is trained using experimental data to achieve the correlation between the amount of PCA and a given particle size under a particular milling duration, i.e., $\text{PCA amount} = f(\text{particle size, milling duration})$. The testing results show that the trained networks have a fairly good generalization capability © 1999 Elsevier Science S.A. All rights reserved.

Keywords: Mechanical milling; Process control agent; Particle size; Back-propagation neural network

1. Introduction

Powder particles in the ball mill are subjected to high-energy collision, which causes the powder particles to be cold-welded together and fractured [1–3]. The cold welding and fracturing process enables powder particles to be always in contact with each other with atomically clean surfaces and with minimized diffusion distance. The essential condition for a successful mechanical alloying process is the balance between cold welding and fracturing. However, this balance in most cases may not be obtainable by the milling process itself, especially if soft materials are used. For such cases, cold welding among powder particles, and between powder particles and milling tools (bowl and balls), becomes a serious problem. The degree of cold welding is dependent on the ductility and the ability to cold welding of the powder to be milled. Depending upon which process is dominant during mechanical alloying, i.e. micro-forging or fracturing, powder particles may grow in size through agglomeration by cold welding, and may change from equi-axed particles to platelet or flake particles by micro-forging or become smaller in size through the fracture process. In order to obtain the balance between the fracturing and welding, a process control agent (PCA) is normally used in the milling

processes [4,5]. The amount of PCA used in a milling process is dependent on the properties of the powder particles, the impact energy and type of PCA.

There exist numerous types of process control agents. Amongst them, stearic acid is one of the most commonly used and effective PCA. The percentage of stearic acid used in a mechanical alloying process is about 1–3 wt% of the powder weight, depending upon the properties of the materials to be milled. For example, for ductile materials, more stearic acid is needed and vice versa. Because the melting temperature of stearic acid is 68°C, it exists in a solid state at the beginning of milling. In this form, the stearic acid may not be homogeneously distributed and hence may result in an inhomogeneous distribution of particle size. Other PCAs include heptane [6], ethyl acetate [7], ethylenebidi-steramide [8,9], polyethylene glycol [10], dodecane [11], hexanes [12,13], methyl alcohol [14] and ethyl alcohol [15], which are also widely used in the milling process. In the present study, two types of material, i.e., Al and Mg, are involved in ball milling with two different kind of PCA. The results are analyzed to find the influence of the PCA on the final particle size after ball milling.

The particle size is a critical parameter in controlling inter-reactions between the different constituents. Although two brittle materials can be mechanically alloyed, one soft material as a binder may assist the rate of reaction. Cold

*Corresponding author. E-mail: mpezyf@nus.edu.sg

welding is an essential requirement of alloying. Without cold welding, the reaction rate is slow. Therefore the particle sizes should be relatively large. However, excessive cold welding between particles may also slow the reaction rate and finally resist the reaction. This is due to no new surfaces being produced by fracturing events, which is an essential process of the balance between the cold welding and fracturing of the powder particles. Since the size of the particles is a function of the amount of PCA, the prediction of the amount of PCA required based on the size of the particles becomes an important issue. Aikin and Courtney [16] applied a discretized form of the fission–fusion equation to the modelling of particle size distributions during mechanical milling. In this paper, neural networks have been used for predicting the amount of PCA required for a particular mean particle size under a particular milling duration.

The experimental procedure is firstly described. The results are then presented and discussed. In order to predict the particle size under different milling conditions, the back-propagation neural networks are trained by using part of the experimental data. Every combination of material (Mg or Al) and PCA has its own neural network. The neural network training process is briefly described. The test results are presented and discussed to assess the neural network's generalization capability.

2. Experimental

Elemental Al and Mg powder particles were milled in a planetary ball mill operated at 250 rpm. Sixteen steel balls of 20 mm diameter are being used in all milling processes. To avoid the influence of oxidation during milling, the milling vial was filled with Ar gas before milling. 0.5–4 wt% of stearic acid, $\text{CH}_3(\text{CH}_2)_{16}\text{CO}_2\text{H}$, or polyethylene glycol, $\text{HO}(\text{C}_2\text{H}_4\text{O})_n\text{H}$, was used in the milling to study the effect of PCA.

After each run, the weight of the powder collected was measured. The mean particle size and distribution of the particles under different milling conditions were measured using two instruments, namely a Horiba LA-910 laser scattering particle size distribution analyzer using a refractive index (in water) of 1.60, and a Quantimet image analyzer.

3. Results and discussion

Fig. 1 shows the change in mean particle size of the ball milled Mg with different percentages of stearic acid at different durations. The size distribution shows that the particle size increases at the initial stage of milling followed by continuous decrease until about 5 h of milling. After 5 h of milling, the size of the milled particles is almost unchangeable. The increase in particle size at the initial

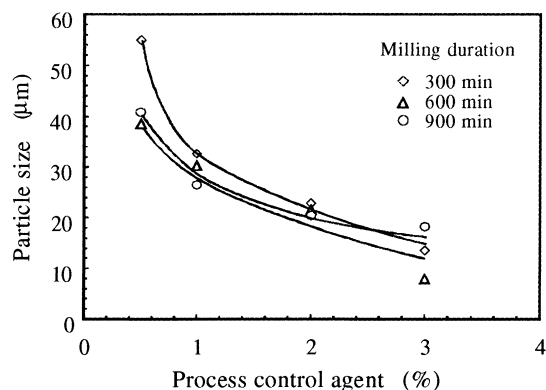


Fig. 1. Mean particle size of Mg as a function of milling duration and the amount of stearic acid.

stage is due to inhomogeneous distribution of the PCA. Due to this uneven distribution of the PCA at the initial stage, Mg particles contact each other without the preventative action of PCA. Cold welding therefore becomes a dominating factor. At longer milling duration, the molten PCA has been homogeneously distributed resulting in an even reduction in particle size. It can be seen that increase in PCA has led to a reduction in particle size.

Fig. 2 shows the change in the weight fraction of Mg recovered after milling with different percentages of stearic acid at different milling durations. Cold welding may take place between the powder particles as well as between the milling tools and the powder particles. Two trends can be found in Fig. 2, namely that the powder recovered increases with an increase in the PCA, and slightly longer milling time may lead to an increase in the powder recovered. It is obvious that an increase in the PCA may resist cold welding between the particles and hence result in fracturing of the powders. Although the PCA will be exhausted with prolonged milling, for a short milling duration, the homogeneity of the PCA distribution is essential. When the milling duration is too short, the portion of powder without PCA may be cold welded onto the surfaces of the milling tools, which leads to less recovery of powder.

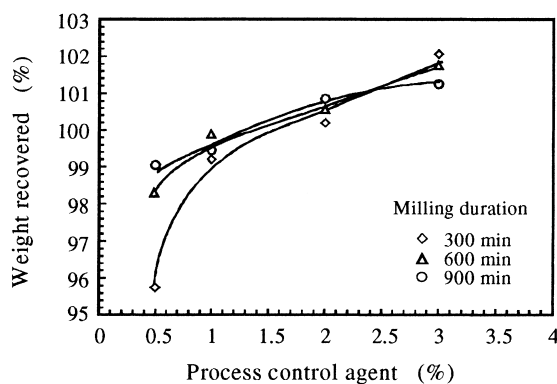


Fig. 2. Weight fraction of milled Mg for various durations with different amounts of stearic acid.

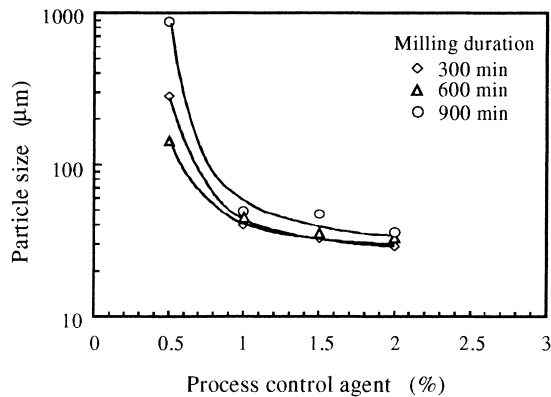


Fig. 3. Mean particle size of Mg as a function of milling duration and the amount of polyethylene glycol.

Fig. 3 shows the mean particle size of the ball milled Mg with different percentages of polyethylene glycol as PCA at different milling durations. A similar trend as for Mg with stearic acid can be observed, i.e. that the particle size decreases with increase in the amount of PCA. However, polyethylene glycol does not reduce the particle size as effectively as stearic acid, relatively high percentages of polyethylene glycol having to be used.

Fig. 4 shows the mean particle size of the ball milled Al with different percentages of stearic acid as PCA at different milling durations. An increase of particle size in the initial stage can be observed. A different trend is found than for the milling of Al powder in that the Al particle size increases with milling duration after 5 h of milling if the stearic acid concentration is below 1 wt%. If the stearic acid is above 2 wt%, the particle size of the Al becomes small. The formation of large Al particles is due to the nature of the fcc structure of Al. Since fcc Al has more slip systems, it is easily deformed and cold welded together, leading to the formation of large particle sizes. When 0.5 wt% of stearic acid was used, particle sizes of about 3–5 mm were obtained. It was interesting to find that a small ball was enclosed within a big ball.

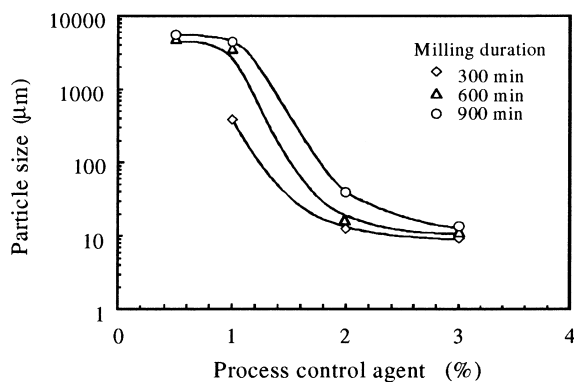


Fig. 4. Mean particle size of Al as a function of milling duration and the amount of stearic acid.

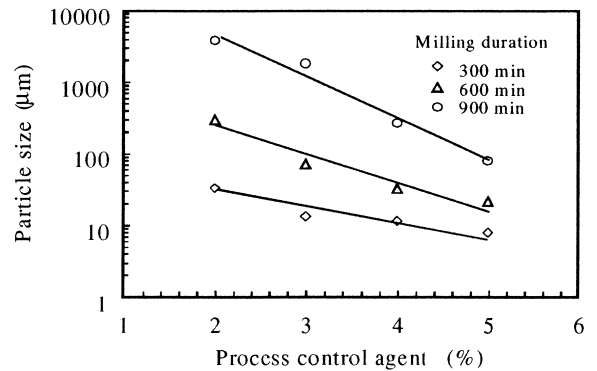


Fig. 5. Mean particle size of Al as a function of milling duration and the amount of polyethylene glycol.

Fig. 5 shows the mean particle size of the ball milled Al with different percentages of polyethylene glycol as PCA, for different milling durations. It is observed that polyethylene glycol is not as effective as stearic acid for particle size reduction. It was also observed that very large particle sizes were resulted when the PCA amount was below 2 wt%. This is because too much cold welding leads not only to dramatic increase in particle size but also to loss of powder due to cold welding with the milling tools. Moreover, polyethylene glycol is not as stable as stearic acid, since a fast increase in particle size with longer milling duration.

4. Prediction of the amount of PCA for a given particle size under a particular milling duration

4.1. The back-propagation (BP) neural network

Based on the experimental data, it is clear that for a given base material and PCA type, the relationship between the PCA amount, particle size, and duration is highly non-linear. One common approach to approximate this kind of relationships is the regression method. However, since the nature of the non-linearity relationships is not known, it must be assumed first. The neural network is typically a “black-box” approach to model unknown relationships based on observed input–output data. In other words, there is no need to know the nature of the non-linearity relationship. In this paper, the back-propagation neural network is employed to learn the relationships between the PCA amount, the particle size, and the milling duration.

Neural networks (NNs) are an information processing technique that simulates biological neurons using computers. One of the most important application of NN is modelling a system with an unknown input–output relation. Usually, accurate information of the system is not available and it is possible only to utilize a number of examples observed from the actual system, called the training set. Given a fixed architecture of the network, learning is carried out through modifying the parameters that eventually mini-

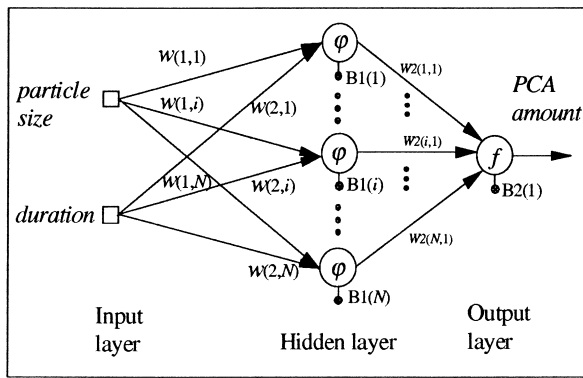


Fig. 6. The BP neural network for approximating the function: $PCA\text{-}amount=f(\text{particle-size, duration})$.

minimize a certain loss function. Amongst various NN architectures, the back-propagation (BP) neural network is a common method that uses the Delta Rule for learning and approximating any non-linear functions [17]. The construction of a BP-network involves one input layer (sensory units), one output layer (response) and one or more hidden layers. Fig. 6 shows the three-layer BP-network used in this study. The transformation from the input space to the output space can be mathematically expressed as:

$$F(\mathbf{x}) = f(\mathbf{W2} \times \phi(\mathbf{W} \times \mathbf{x} + \mathbf{B1}) + \mathbf{B2}), \quad (1)$$

where f and ϕ are transfer functions that are usually taken as sigmoidal functions. \mathbf{W} and $\mathbf{W2}$ are weights vectors associated with the hidden layer and the output layer, respectively; $\mathbf{B1}$ and $\mathbf{B2}$ the bias vectors associated with the hidden layer and output layer, respectively; and \mathbf{x} is the input vector. Given a number of known input and targeted output pairs, the network firstly uses randomly generated weights and biases to produce a set of outputs. The sum-squared error between the actual outputs and the targeted outputs is then calculated. The network changes the values of weights and biases in the direction of steepest descent with respect to this error. This process is repeated until the sum-squared error is minimized. This is called the BP learning procedure. Trained BP-networks tend to give reasonable answers when presented with inputs that they have never seen. This generalization property makes it possible to train a network on a representative set of input/output pairs and then obtain good results for new input without training the network on all possible input/output pairs.

As shown in Fig. 6, the BP-network used in this study has three layers. There are two input neurons in the input layer, i.e., particle size and duration. The output layer has only one neuron, which represents the PCA amount. A single hidden layer is selected. Based on the rules for BP architecture selection proposed by Carpenter and Hoffman [18], the number of hidden neurons is calculated based on the following:

$$N \geq 4M + 1, \quad (2)$$

where N is the total number of training pairs and M is the

number of hidden neurons. However, M can only be used as an initial value to start with. The actual number of hidden neurons has to be obtained through trial-and-error.

The transfer functions for both the hidden layer and the output layer are chosen as Log-sigmoid function. The neural network program is written using the MATLAB Neural Network Toolbox [19].

Since the error used for BP learning rule is the sum-squared error for all of the training pairs, the selection of the error goal for network training must be based on the training pairs. In this study, an expected percentage error goal (error%) for each training pairs is assumed. The expected sum-squared error goal (SSE-goal) is then calculated as follows:

$$SSE\text{-}goal = \sum_{i=1}^N (T_i \times \text{error}\%)^2, \quad (3)$$

where T_i is the target output of any training pair and N is the total number of training pairs. To avoid over-fitting the training samples, the error% is taken to be around 10%.

4.2. The training and testing results

Three BP-networks have been trained for three combinations of base material and PCA, i.e., Mg with stearic acid, Mg with polyethylene glycol, and Al with stearic acid. The training and testing results are presented in the following sections.

4.2.1. Mg with stearic acid as PCA

The experimental data for the training and testing of the network are shown in Table 1. There are totally 16 pairs of data (columns 2–4). Amongst them, four pairs (shaded in the table) are to be used for testing the network, whilst the

Table 1

Experiment data and predicted output from the BP-network for Mg with stearic acid (the shaded data are testing samples that have not been used in BP-network training)

PCA (wt%) predicted (NN)	PCA (wt%) actual	Duration (h)	Mean particle size (µm)
0.47%	0.5%	5	55.0
0.48%	0.5%	10	38.6
0.53%	0.5%	15	40.8
0.54%	0.5%	20	50.7
0.88%	1.0%	5	32.7
1.13%	1.0%	10	30.3
1.19%	1.0%	15	26.4
1.02%	1.0%	20	27.5
1.96%	2.0%	5	22.8
1.97%	2.0%	10	21.7
2.17%	2.0%	15	20.4
2.11%	2.0%	20	23.2
2.84%	3.0%	5	13.6
2.71%	3.0%	10	8.0
2.62%	3.0%	15	18.1
3.31%	3.0%	20	18.6

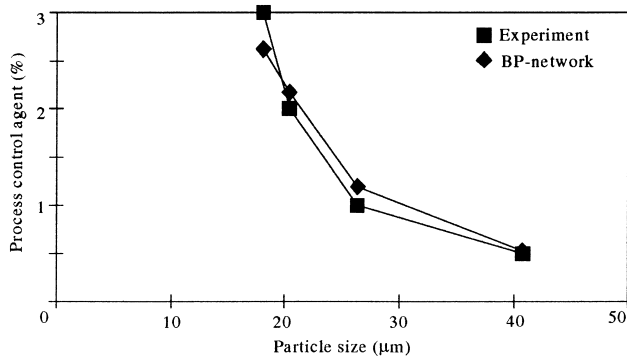


Fig. 7. Experiment data versus BP-network output (Mg with stearic acid).

remaining 12 pairs are used for training. The final network structure has five hidden neurons. With the trained network, the PCA amount for all the training and testing pairs can be predicted as shown in column 1 of Table 1. Compared with the actual PCA amount, the largest relative predicting errors are 13% and 19% for the training pairs and testing pairs, respectively. The comparison between the network output and the experimental data for the testing samples is depicted in Fig. 7, which clearly indicates that a good agreement has been achieved.

4.2.2. Mg with polyethylene glycol as PCA

The experimental data for the training and testing of the network are shown in Table 2. There are totally 16 pairs of data (columns 2–4). Amongst them, four pairs (shaded in the table) are to be used for testing the network, whilst the remaining 12 pairs are used for training. The final network structure has seven hidden neurons. With the trained network, the PCA amount for all of the training and testing pairs can be predicted as shown in column 1 of Table 2. Compared with the actual PCA amount, the largest prediction errors are

Table 2

Experiment data and predicted output from the BP-network for Mg with polyethylene glycol (the shaded data are testing samples that have not been used in BP-network training)

PCA (wt%) predicated (NN)	PCA (wt%) actual	Duration (h)	Mean particle size (µm)
0.47%	0.5%	5	281.0
0.53%	0.5%	10	143.6
0.39%	0.5%	15	878.0
0.40%	0.5%	20	1624.0
1.20%	1.0%	5	39.6
1.24%	1.0%	10	45.0
1.28%	1.0%	15	51.6
1.14%	1.0%	20	66.6
1.51%	1.5%	5	33.7
1.59%	1.5%	10	34.2
1.44%	1.5%	15	47.5
1.59%	1.5%	20	43.8
1.76%	2.0%	5	28.6
2.15%	2.0%	10	33.4
1.78%	2.0%	15	34.6
1.90%	2.0%	20	35.6

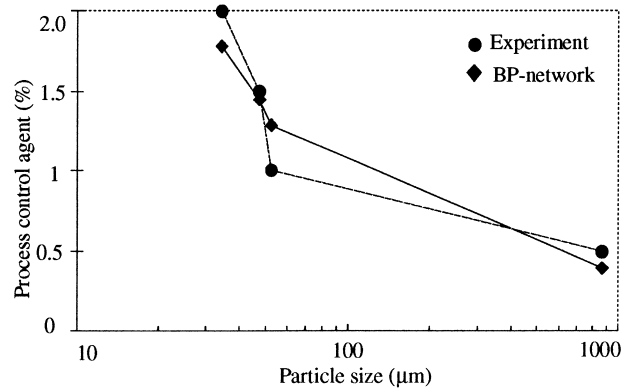


Fig. 8. Experiment data versus BP-network output (Mg with polyethylene glycol).

24% and 28% for the training pairs and testing pairs, respectively. The comparison between the network output and the experimental data for the testing samples is depicted in Fig. 8, which indicates that a fair agreement has been achieved.

4.2.3. Al with stearic acid as PCA

The experimental data for the training and testing of the network are shown in Table 3. There are totally 16 pairs of data (columns 2–4). Amongst them, four pairs (shaded in the table) are to be used for testing the network, whilst the remaining 12 pairs are used for training. Through trial-and-error, the final network structure has eight hidden neurons. With the trained network, the PCA amount for all of the training and testing pairs can be predicted as shown in column 1 of Table 3. Compared with the actual PCA amount, the largest prediction errors are 34% and 40% for the training pairs and testing pairs, respectively. The comparison between the network output and the experimen-

Table 3

Experiment data and predicted output from the BP-network for Al with stearic acid (the shaded data are testing samples that have not been used in BP-network training)

PCA (wt%) predicated (NN)	PCA (wt%) actual	Duration (h)	Mean particle size(µm)
0.53%	0.5%	5	3018
0.70%	0.5%	10	4719
0.67%	0.5%	15	5286
0.51%	0.5%	20	5076
0.94%	1.0%	5	375
0.94%	1.0%	10	3979
0.89%	1.0%	15	4293
0.97%	1.0%	20	3436
2.15%	2.0%	5	13.5
1.99%	2.0%	10	13.4
2.10%	2.0%	15	38.4
2.35%	2.0%	20	11.9
2.93%	3.0%	5	9.3
2.73%	3.0%	10	10.9
2.83%	3.0%	15	13.4
2.71%	3.0%	20	12.1

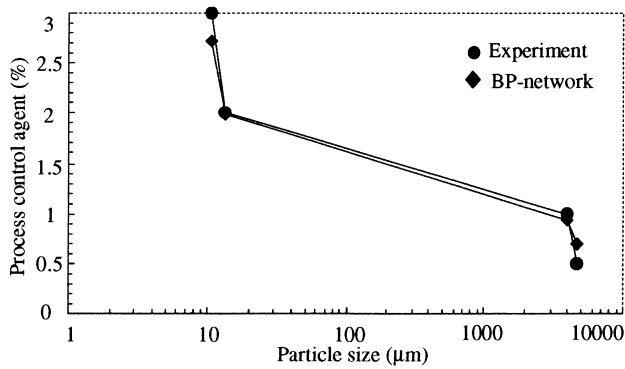


Fig. 9. Experiment data versus BP-network output (Al with stearic acid).

tal data for the testing samples is depicted in Fig. 9, which clearly indicates that a good agreement has been achieved.

In summary, the comparison between the BP-network's predictions and the experiment data for the three base material and PCA combinations clearly shows that, to a fairly good accuracy, the BP-network is able to predict the amount of PCA required in order to achieve a certain mean particle size under a particular milling duration. This suggests that when an accurate mathematical model is not available, as at present, the neural network model can be used to guide the ball milling process in terms of predicting the amount of PCA required. The deviation between the BP-network predictions and the experiment data could be attributed to the following:

1. The limited number of training samples leads to being certain parts of the actual function curve not being learnt, due to lack of information.
2. The particle size distributions of some experimental data are not of normal distribution. This could lead to noise in the experiment data (mean particle size).
3. The BP-network training is essentially a trial and error process, although there are certain guidelines. This suggests that there is no certainty that the final network structure is the optimal.

Therefore, the BP-networks can be improved with more experiment data collected, and more training carried out.

5. Conclusions

In this paper, the authors have presented experimental results on the mechanical milling of Mg and Al with different process control agents (PCA), i.e., stearic acid and polyethylene glycol. The influence of the amount of PCA on the resulting mean particle size was analyzed and discussed. It was found that there is a critical amount of PCA for a particular powder material, below which the particle size tends to increase and above which it tends to decrease.

The final particle size is a result of the PCA type, the PCA amount, and the milling duration. In order to achieve a particular particle size under a particular milling duration, BP-networks have been applied to predict the amount of PCA required. Three BP-networks were constructed and trained for the combinations of (Mg, stearic acid), (Mg, polyethylene glycol), and (Al, stearic acid). The results showed a fairly good agreement between BP-network predictions and the experiment data. Although some deviations exist from the experiment data, this approach appears to be valid.

References

- [1] J.S. Benjamin, Powder metallurgy, in: Proceedings of the 1992 Powder Metallurgy World Congress, San Francisco, CA, 21–26 June 1992, Publ. Metal Powder Industries, vol. 7, 1992, p. 155.
- [2] J.S. Benjamin, Mater. Sci. Forum 88(90) (1992) 1.
- [3] J.S. Benjamin, in: E. Arzt, L. Schultz (Eds.), DGM Confer., Calw-Hirsau (FRG), October 1988, Informationsgesellschaft Verlag, p. 3.
- [4] P.S. Gilman, J.S. Benjamin, Ann. Rev. Mater. Sci. 13 (1983) 279.
- [5] W.Y. Lim, M. Hida, A. Sakakibara, Y. Takemoto, S. Yokomizo, J. Mater. Sci. 28 (1993) 3463.
- [6] T.S. Suzuki, M. Nagumo, Mater. Sci. Forum 179 180 181 (1955) 189.
- [7] X.P. Xiu, Ph.D. Thesis at KULeuven, Belgium, (1991) 34.
- [8] P.S. Gilman, W.D. Nix, Metall. Trans. A 12A (1981) 813.
- [9] A. Arias, Chemical Reactions of Metal Powders with Organic and Inorganic Liquids during Ball Milling, NASA TN D-8015 (1975).
- [10] L. Lu, M.O. Lai, S. Zhang, Key Eng. Mater. 104 105 106 107 (1995) 111.
- [11] A.M. Harris, G.B. Schaffer, N.W. Page, in: J.J. deBarbadillo, F.H. Froes, R. Schwarz (Eds.), Proceedings of the Second International Conference on Structural Applications of Mechanical Alloying, Vancouver, BC, 20–22 September 1993, ASM Intern. Mat. Park, OH, p. 15.
- [12] R.B. Schwarz, P.B. Desch, S.R. Srinivasan, in: J.J. deBarbadillo, F.H. Froes, R. Schwarz (Eds.), Proceedings of the Second International Conference on Structural Applications of Mechanical Alloying, Vancouver, BC, 20–22 September 1993, ASM Intern. Mat. Park, OH, p. 227.
- [13] J.R. Groza, M.J.H. Tracy, in: J.J. deBarbadillo, F.H. Froes, R. Schwarz (Eds.), Proceedings of the Second International Conference on Structural Applications of Mechanical Alloying, Vancouver, BC, 20–22 September 1993, ASM Intern. Mat. Park, OH, p. 327.
- [14] M. Umamoto, T. Itsukaichi, J. Cabanas-Moreno, I. Okane, in: J.J. deBarbadillo, F.H. Froes, R. Schwarz (Eds.), Proceedings of the Second International Conference on Structural Applications of Mechanical Alloying, Vancouver, BC, 20–22 September 1993, ASM Intern. Mat. Park, OH, p. 245.
- [15] A. Malchere, E. Gaffet, in: J.J. deBarbadillo, F.H. Froes, R. Schwarz (Eds.), Proceedings of the Second International Conference on Structural Applications of Mechanical Alloying, Vancouver, BC, 20–22 September 1993, ASM Intern. Mat. Park, OH, p. 297.
- [16] B.J.M. Aikin, T.H. Courtney, Metal. Trans. A 24A (1993) 2465.
- [17] R. Beale, T. Jackson, Neural Computing – An Introduction, Adam Hilger, Bristol, 1990.
- [18] W.C. Carpenter, M.E. Hoffman, Art. Intell. Eng. Des. Anal. Manuf. 11 (1993) 33.
- [19] The MathWorks, Neural Network Toolbox for Use with MATLAB (1993).